



Shell cracking strength in almond (*Prunus dulcis* [Mill.] D.A. Webb.) and its implication in uses as a value-added product

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Abstract

Researchers are currently developing new value-added uses for almond shells, an abundant agricultural by-product. Almond varieties are distinguished by processors as being either hard or soft shelled, but these two broad classes of almond also exhibit varietal diversity in shell morphology and physical characters. By defining more precisely the physical and chemical characteristics of almond shells from different varieties, researchers will better understand which specific shell types are best suited for specific industrial processes. Eight diverse almond accessions were evaluated in two consecutive harvest seasons for nut and kernel weight, kernel percentage and shell cracking strength. Shell bulk density was evaluated in a separate year. Harvest year by almond accession interactions were highly significant ($p \leq 0.01$) for each of the analyzed variables. Significant ($p \leq 0.01$) correlations were noted for average nut weight with kernel weight, kernel percentage and shell cracking strength. A significant ($p \leq 0.01$) negative correlation for shell cracking strength with kernel percentage was noted. In some cases shell cracking strength was independent of the kernel percentage which suggests that either variety compositional differences or shell morphology affect the shell cracking strength. The varietal characterization of almond shell materials will assist in determining the best value-added uses for this abundant agricultural by-product.

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1. Introduction

Almonds are a crop of major importance throughout the world's temperate growing regions with worldwide almond production in 2005 being approximately 1.65 million ton from a total of 1.8 million ha (FAOSTAT, 2006). Almond kernels are nutritious for humans and the outer hulls have been used as a palatable carbohydrate source for dairy cattle. Profits derived from the sale of hulls to feedlots partially offset the cost of hulling and shelling the almond crop (DePeters et al., 2000).

Between the outer hull and the nutritious kernel lies the shell, primarily composed of cellulose, hemicellulose and lignin. Shell strength is a function of the proportion of

these chemicals, shell morphology, fiber content and the outer shell adherence. The shell varies in cracking strength between varieties, and terms such as 'papershell' and 'extreme hardshell' are used as descriptors for the shell types of various varieties (Gülcan, 1985). The kernel percentage (kernel weight/nut weight) is used as an indicator of relative shell strength and this value provides almond handlers with a general guide of the almond variety's shell cracking requirements. However, two almond accessions having a similar kernel percentage might have different shell strengths, and processing a soft shell almond on a cracking line dedicated to hard shell almonds would lead to significant kernel damage. Shell strength is highly heritable (Kester et al., 1977), with hardshell dominant over soft or papershell (Grasselly, 1972).

When delivered at the processing facility, the uniformity of a given almond lot is very important to the grower in

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order to obtain a maximum price from the crop. Growers are motivated to keep almond varieties separate since varietal mixing in the orchard at harvest or during the processing operation leads to product non-uniformity (Anon, 1997), and growers are penalized financially for the mixed lot. Almond hulling/cracking facilities are capable of processing each lot separately to ensure varietal uniformity of the edible product, and also providing large volumes of clean and uniform shells to be available for specific industrial uses.

There are several proposed uses for almond shells where varietal uniformity is not a critical issue, and the characterization of the shell material is of probable little importance. Large volumes of shells are currently consumed in co-generation plants for direct production of electricity. Shells have also been utilized as organic inclusions in porous ceramic bodies for tissue engineering and drug delivery systems applications (Rodriguez-Lorenzo and Ferreira, 2004), as an additive to 'driller's mud' to reduce the chance of equipment sticking during the drilling of new wells (Mayeux and Ruby, 1999) and as a substrate in soilless or hydroponic culture (Lao and Jimenez, 2004).

Some researchers are focusing on the valuable shell by-products obtained after flash pyrolysis, gasification or ozonation (Font et al., 1988; Gonz  les et al., 2002; Mitchell et al., 2003). There is keen interest in using this high volume by-product of the almond industry as a source of granular activated carbons for removing heavy metals, organic contaminants and chloride compounds from drinking water and industrial wastewaters (Ahmedna et al., 2004). Similarly, other types of nutshells have been evaluated as activated carbon feedstocks for their ability to adsorb contaminants (Wartelle and Marshall, 2001). Wide differences were found in the specific adsorptive ability of activated carbons produced from various nutshells. Adsorptive capacity of activated carbons relates closely with the carbon's pore surface area and relative distribution of pore volumes (Balci et al., 1994). Clearly both nutshell feedstock source and activation conditions directly affect the specific properties of activated carbons derived thereof (Toles et al., 2000), and nutshell sources must be both defined and uniform in order to precisely determine the adsorptive properties of activated carbons derived from them. Currently, aside from producing and actual testing of activated carbons from defined nutshell sources, there are no known predictive nutshell characteristics or properties that are useful in determining the adsorptive capacities of carbons produced from them. In contrast to activated carbon production from nutshell feedstocks, ground almond shells have been used successfully to adsorb pentachlorophenol from contaminated wastewaters. With a much lower internal surface area as compared with activated carbons, adsorption by the shell particles was attributed to functional groups in the shell material that facilitated bonding of the pentachlorophenol with the shells (Esteveinho et al., 2006).

Given the interest in the utilization of almond shells as a value-added product, and the fact that different nutshell sources have been shown to vary widely in their favored industrial uses, it is important to characterize the physical and chemical traits of important almond varieties. Varietal characterization of almond shells would assist in identifying which shell types are best suited for any particular industrial application. Furthermore, characterization can elucidate whether or not any specific physical trait or property correlates well with performance in a given industrial application. The objective of this present study was to examine the variability of eight almond accessions over two harvest seasons in terms of nut and kernel weight, kernel percentage and shell cracking strength. The relationship between shell strength and the other parameters was also examined.

2. Methods

2.1. Plant materials

Almonds were obtained from trees grown at the San Joaquin Valley Agricultural Sciences Center in Parlier, California. Samples of Mission, Monterey, Padre, Ruby, Targona and Tuono were obtained from 12 to 14 year old trees propagated on Nemaguard peach rootstock, with two trees available for each cultivar. The remaining two accessions, Y113-20 and Y120-70, were own rooted trees in their fifth leaf during the first year of the study.

In each study year, almond samples were collected after hull split during the normal period of commercial harvest. The nuts were knocked from the trees and air dried on tarps on the orchard floor. Subsamples were collected from the bulk harvests on tarps and frozen before processing. The air dried samples were then oven-dried overnight at 40 °C prior to analysis.

2.2. Sample analysis

De-hulled almond fruit (nuts) were weighed prior to an analysis of shell cracking strength using an Instron, Series IX Automated Materials Testing System, Model 4201 (Canton, MA, USA). The Instron was equipped with a 1 kN Static Load Cell. Individual nuts were placed on a stationary metal plate at the base of the Instron such that the suture plane of the nut was parallel to the metal plate, and perpendicular to the direction of compression. Compression of the nut was then begun from above, with the Instron's crosshead speed descending at a rate of 25.0 mm/min. The maximum force necessary to break the shell was recorded. Twenty-five individual nuts of each almond accession were analyzed each year. Kernels from each nut were then weighed after cracking and kernel percentage was calculated for each nut (kernel wt/nut weight).

To determine bulk density, oven dried shells were pulsed repeatedly in a Krups type 208 mill (Medford, MA, USA) and sieved to achieve a particle size between 1 and 2 mm. A

2.0 cm³ sample (tapped) of shell particulates was then weighed on an analytical balance. There were seven replications per accession, using shells from the 2007 crop year.

2.3. Statistical analyses

Factors ‘Harvest Year’ and ‘Almond Accession’ were used as independent effects in a two-way analysis of variance (ANOVA). There were two harvest years (2004 and 2005) and eight almond accessions (Tuono, Tarragona, Ruby, Padre, Mission, Monterey, Y113-20 and Y120-70) used in this study. For these analyses, twenty five nuts were used from each accession in each harvest year. Shell bulk density was analyzed separately in a one-way ANOVA to compare shell material densities of the eight almond accessions in a single harvest year. In this analysis, there were seven replications per accession. Prior to any analyses, all data were examined for homogeneity of variance, and no data transformations were found necessary to satisfy ANOVA requirements. If a significant *F*-value ($p \leq 0.01$) was obtained from the ANOVA table, Tukey’s Honestly Significant Difference test was used to compare differences between means. Pearson correlation coefficients were calculated for all pairs of dependent variables from the two-way ANOVA raw data file ($n = 400$). Data presented in this study were analyzed and tabulated with JMP stats (Cary, NC, SAS, Version 7.0).

3. Results and discussion

Global almond production has increased steadily at a rate of five percent annually since the early 1990s with major new production coming from both North and South American as well as Australian orchards (USDA Foreign Agricultural Service, 2004). The increased kernel production comes with an associated increase in almond shells from processors. But while numerous studies have examined varietal variability in almond kernels with respect to oil content, amino acid composition, and both sugar and fiber content (Cordeiro et al., 2001; Garcia-López et al., 1996; Lansari et al., 1994; Saura Calixto et al., 1981), specific studies on varietal variability of almond shells have been much fewer. Askin et al. (2007) observed significant differences in almond shell thickness among 26 sampled Turkish almond genotypes and determined that kernel oleic acid content correlated positively with shell thickness. Breeding efficiency for an important horticultural trait can be improved by identifying and utilizing significant correlations between simple physical and/or chemical characters and the important horticultural trait of interest. Similarly, it is desirable to identify specific physical or chemical traits associated with almond shells that correlated significantly with their ability to be utilized in particular industrial applications.

In the current study, harvest year by almond accession interaction was observed to be significant ($p \leq 0.01$) for nut and kernel weights, kernel percentage and shell crack-

ing strength. Furthermore, all main effects were significant ($p \leq 0.01$) except for the effect of harvest year on shell cracking strength (490.0 N in 2004 vs. 486.8 N in 2005), which was not significant. Shell bulk density was affected significantly ($p \leq 0.01$) by almond accession (Table 1).

Nut and kernel weights as well as kernel percentage were significantly ($p \leq 0.01$) higher in 2005 as compared with the previous year. Averaged across accessions, nut weight, kernel weight and kernel percentage values were 2.93 g, 1.09 g and 40.36%, respectively in 2005, whereas in 2004 these values were 2.62 g, 0.91 g and 37.11%, respectively (Table 2). Averaged across accessions, nut weights ranged significantly ($p \leq 0.01$) from 4.11 g (Y113-20) to 1.68 g (Padre). Compared to nut weights, kernel weights were more similar among the evaluated almond accessions, with Tuono, Monterey, Y113-20 and Y120-70 not differing significantly in kernel weight. Kernel weight of accession Padre was numerically lowest (0.89 g). Kernel percentage ranged significantly ($p \leq 0.01$) from 26.00% (Y113-20) to a high of 53.05% (Padre). Shell bulk density of Y113-20 was significantly ($p \leq 0.01$) higher than all other examined almond accessions, and bulk density of Padre was significantly ($p \leq 0.01$) lower. Shell bulk density ranged significantly ($p \leq 0.01$) from 0.476 to 0.685 g/cm³ for the evaluated almond accessions, somewhat higher than the 0.37 g/cm³ value provided by Wartelle and Marshall (2001) for almond shells of an unspecified variety. Shell cracking strength of Monterey was significantly ($p \leq 0.01$) weaker than the other seven accessions, and shell strength was significantly ($p \leq 0.01$) highest in Y113-20. Averaged across almond accessions, shell strength was not influenced significantly by harvest year.

While shell cracking strength varied considerably between almond accessions, all accessions used in this study except for Monterey are considered by the industry to be “hard” shell almonds. This distinction is important to almond processors, as textured steel rollers are needed to crack hard shell varieties as compared to the solid rubber rollers used to crack the softer shell varieties. Shell strength values presented in Table 2 may be somewhat predictive in determining the shell strength threshold at which rubber rollers can no longer be used. Since accession Monterey can be cracked adequately on rubber rollers but steel

Table 1
ANOVA of nut weight, kernel weight, kernel percentage, shell strength and shell bulk density as a function of two harvest years and eight almond accessions

	df	Mean square				
		Nut wt	Kernel wt	Kernel %	Shell strength	Bulk density
Year	1	9.38**	3.22**	1053.33**	10.99	–
Accession	7	33.77**	0.32**	4835.93**	20747.40**	0.09**
<i>Y</i> × <i>A</i>	7	8.87**	0.39**	343.20**	2885.52**	–
Error	384	0.11	0.013	6.15	37.07	0.000151 (df = 48)

** Significant at $p < 0.01$.

Table 2

Nut and kernel weights, kernel percentage, shell bulk density and shell cracking strength of eight almond accessions sampled in 2004 and 2005

Factor	Nut wt (g)	Kernel wt (g)	Kernel %	Bulk density (g/cm ³) ^a	Shell strength (N)
<i>Year</i>					
2004	2.62 ± 0.76 ^b	0.91 ± 0.15	37.11 ± 9.89	–	490.0 ± 202.0
2005	2.93 ± 1.07	1.09 ± 0.17	40.36 ± 9.82	–	486.8 ± 214.5
<i>Accession</i>					
Tuono	3.47 ± 0.52 b ^c	1.10 ± 0.13 a	32.01 ± 4.23 d	0.544 ± 0.006 d	633.1 ± 66.1 b
Tarragona	3.08 ± 0.67 c	0.95 ± 0.17 cd	31.01 ± 2.54 d	0.614 ± 0.009 c	498.7 ± 153.3 cd
Ruby	2.13 ± 0.19 d	0.98 ± 0.09 bcd	46.10 ± 3.10 b	0.497 ± 0.008 e	367.0 ± 58.8 e
Padre	1.68 ± 0.22 e	0.89 ± 0.11 d	53.05 ± 1.80 a	0.476 ± 0.006 f	334.8 ± 55.1 e
Mission	2.18 ± 0.26 d	0.93 ± 0.11 d	42.87 ± 2.49 c	0.645 ± 0.010 b	447.2 ± 74.8 d
Monterey	2.34 ± 0.28 d	1.10 ± 0.13 a	47.57 ± 5.43 b	0.500 ± 0.007 e	227.9 ± 53.4 f
Y113-20	4.11 ± 1.09 a	1.06 ± 0.27 ab	26.00 ± 1.51 e	0.685 ± 0.011 a	874.4 ± 60.4 a
Y120-70	3.20 ± 0.43 bc	1.00 ± 0.25 abc	31.28 ± 6.60 d	0.544 ± 0.005 d	524.2 ± 145.3 c

^a Values represent observations ($n = 7$) taken in a single harvest season. Estimates based on a tapped sample.^b Mean ± standard deviation. Values based on 200 observations.^c Values followed by the same letter within a column of almond accessions do not differ significantly at the $p < 0.01$ level according to a Tukey's Honestly Significant Difference Test. Values based on 50 observations.

rollers are necessary for cracking Padre, the maximum shell strength for successful almond cracking on rubber rollers is estimated at approximately 250–300 N. High volume California varieties such as Carmel, Nonpareil and Sonora, whose shells are much softer than Monterey, are unsuitable for strength testing using these described procedures. In preliminary tests with these soft shelled varieties, no precise release of compression force could be noted to indicate when the shell cracked. Shells of these varieties deformed during compression prior to breaking. This would indicate a shell with more fibers and less bony material, as compared with the almond accessions utilized in this study. Thus, these softer shelled almond varieties were not examined in this study.

Although major tonnage varieties Nonpareil and Carmel account for over 50% of all California almond tonnage, varieties Mission, Monterey, Padre and Ruby still contribute significantly to the total California almond crop. Combined inshell harvest of these four varieties in 2006 accounted for 67,758 ton (13.34%) of the total 507,605 ton harvest (Anon, 2007). Based on the calculated kernel percentages in this study, these varieties alone produced over 35,000 ton of almond shells in 2006, or nearly twice the tonnage of shells that are directly burnt after processing in Spain (Pou-Llinas et al., 1990).

Comparing almond accessions from the two harvest seasons, only Ruby, Mission and Y120-70 did not differ significantly between harvest years for nut weight. Almond accessions Tarragona, Padre and Y113-20 had significantly higher ($p \leq 0.01$) nut weights in harvest year 2005 as compared with 2004, and nut weights of Tuono and Monterey were significantly higher ($p \leq 0.01$) in 2004 (Fig. 1a). For kernel weights, no significant difference between harvest years was noted for almond accessions Tuono, Ruby or Monterey (Fig. 1b). Almond accessions Ruby, Padre, Mission and Y113-20 did not differ significantly between harvest years for kernel percentage. Contrasting these accessions were Tuono, Monterey and Y120-70, whose ker-

nel percentages were significantly ($p \leq 0.01$) higher in 2005 (Fig. 1c). Mission was the only almond accession that did not differ significantly between harvest years for shell cracking strength. Shell cracking strength was significantly higher ($p \leq 0.01$) in 2004 for accessions Tuono, Ruby, Monterey and Y120-70, and significantly higher ($p \leq 0.01$) for Tarragona, Padre and Y113-20 in harvest year 2005 (Fig. 1d).

Environmental conditions and orchard cultural practices can be responsible for annual variations in nut characteristics within any given variety. Rain and/or cold weather during the bloom period can reduce bee flights, thereby reducing fruit set, leading to fewer but larger nuts. Kernel and nut size of a given variety is typically larger in years when inclement weather prevailed during the bloom period. Kernel size can be adversely affected by insufficient irrigation during the nut filling period. In this case, a reduced kernel weight would also yield a lower than expected kernel percentage. Insect feeding can also lead to reduced kernel and nut weights, or to nut abortion, depending on the timing of the infestation. Heavy pressure in California almond orchards by the leaf-footed bug (*Leptoglossus clypealis*) during 2006 caused severe damage (reduced yield, shriveled kernels) to the crop throughout the growing region. Overall, kernel percentage was reduced in the affected orchards, and average kernel weights declined as well.

The correlation analysis demonstrates a significant ($p \leq 0.01$) positive link between nut and kernel weight (Table 3), and was in general agreement with previously conducted studies in tree nuts (Hardner et al., 2001; Kester et al., 1977; Rink et al., 1997; Thompson and Baker, 1993). A similar significant positive correlation ($r^2 = 0.567$ vs. $r^2 = 0.598$) between kernel and nut weights was reported by Talhouk et al. (2000) in a diversity study of 106 Lebanese almond accessions. A non-significant positive correlation between shell strength and kernel weight also mirrored ($r^2 = 0.168$ vs. $r^2 = 0.126$) the results

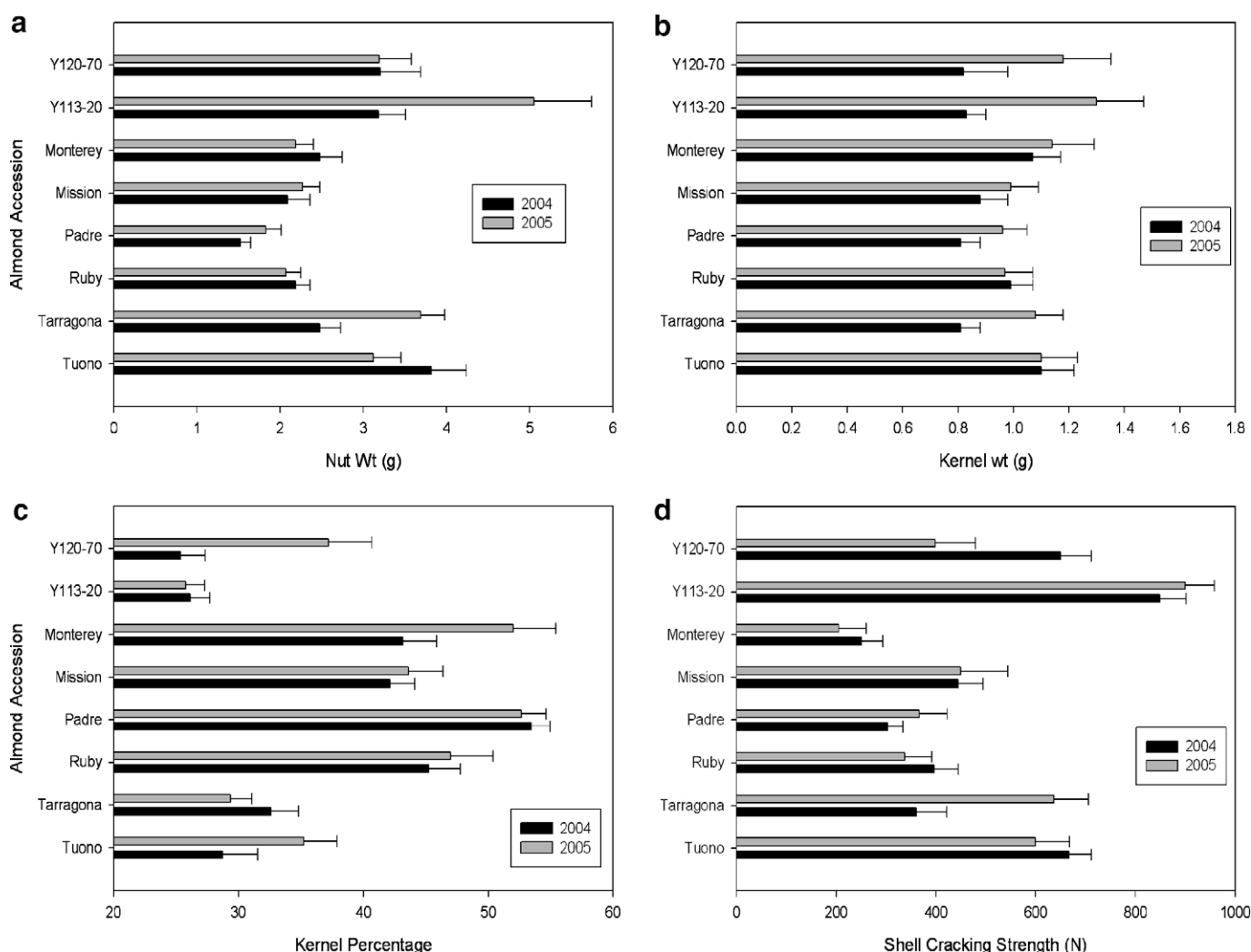


Fig. 1. Almond accession by harvest year values (mean \pm standard deviation) for nut wt (a), kernel wt (b), kernel percentage (c) and shell cracking strength (d). Values are based on 25 observations.

Table 3

Matrix of Pearson correlation coefficients for nut and kernel variables of eight almond accessions analyzed in two harvest years ($n = 400$)

	Nut wt	Kernel wt	Kernel %	Shell strength
Nut wt	1.000			
Kernel wt	0.598**	1.000		
Kernel %	-0.783**	-0.025	1.000	
Shell strength	0.762**	0.126	-0.810**	1.000

** Indicative of a significant ($p < 0.01$) correlation coefficient probability according to a Bonferroni test.

obtained by Talhouk et al. (2000). The current study also determined a significant ($p \leq 0.01$) negative correlation between nut weight and kernel percentage. Kernel weight, on the other hand, did not correlate significantly with kernel percentage. Shell cracking strength was also found to have no significant correlation with kernel weight; however, the highest ($r^2 = -0.810$) significant ($p \leq 0.01$) correlation observed in this study was between kernel percentage and shell cracking strength. Sánchez-Pérez

et al. (2007) calculated a very similar ($r^2 = -0.84$) value for these same variables in an analysis characterizing Western European almond germplasm. These would be considered logical results since as the proportion of almond shell material increases, the shell would generally be more difficult to crack.

There are however, specific results in the current study that are contrary to the significant negative correlation between shell cracking strength and kernel percentage. It is exactly these results that provide evidence for varietal differences in almond shell composition. We observed no significant ($p \leq 0.01$) difference in 2004 between Mission and Monterey in kernel percentage (42.13% vs. 43.14%), but these same almond differed significantly ($p \leq 0.01$) in shell strength (444.9 N vs. 250.1 N). Similarly, kernel percentage of Padre (52.67%) and Monterey (51.99%) did not differ significantly ($p \leq 0.01$) in 2005 whereas the shell cracking strength of these two accessions differed significantly ($p \leq 0.01$) in that year (366.5 N vs. 205.7 N). Almond accessions Y113-20 and Y120-70 did not differ sig-

nificantly ($p \leq 0.01$) for kernel percentage in 2004, but shell cracking strength was significantly ($p \leq 0.01$) greater (849.2 N vs. 650.1 N) for Y113-20 as compared to Y120-70 (Fig. 1c and d). Since kernel percentage is merely a weight ratio, the fact that there are noted differences in shell cracking strength for almond accessions not differing significantly in kernel percentage indicates varietal compositional or structural differences in the almond shells. Differences in shell bulk density between almond accessions do not adequately explain shell cracking strength differences between almonds with similar kernel percentages. While almond accession Y113-20 had the strongest shell type and was significantly ($p \leq 0.01$) highest in shell bulk density, Monterey almond shells, weakest in shell strength of the examined almond accessions, were not the lowest in bulk density.

Some of the proposed value-added uses for almond shells could be greatly affected by the specific types of shells used, and material uniformity could be crucial for specific products or processes. Published estimates of almond shell composition relative to cellulose, hemicellulose and lignin content have reportedly ranged from 29.8 to 50.7%, 19.3 to 29% and 20.4 to 50.7%, respectively (Demirbas, 2002a; Font et al., 1988; Pou-Llinas et al., 1990; Wartelle and Marshall, 2001), indicating varietal variability for these major constituents in the analyzed shells. Recent studies on biomass saccharification for biofuel production has demonstrated that specific lignin structure as well as total lignin content affect biomass enzymatic hydrolysis, and thus the final biofuel yield (Chen and Dixon, 2007; Davison et al., 2006). Biomass lignin content has also been shown to be directly related to the uptake of cadmium (Basso et al., 2004). Research on alternative fuels is becoming increasingly more focused on identifying sources of biomass with high heating values in order to operate more efficiently. A highly significant positive correlation was found between lignin content and higher heating values in 14 sampled biomass fuels (Demirbas, 2001). A higher ash content of the biomass reduced its heating value and an increased extractive content made a given biomass more desirable as a fuel source (Demirbas, 2002b). An assortment of embedded compounds or secondary metabolites can be responsible for increased extractive content in a given biomass, and while their individual heating values may vary, collectively they provide increased heat of combustion per unit weight of biomass as compared with biomass sources low in extractive content. Other researchers are sampling many diverse biomass sources and determining the extent to which the chemical characterization (cellulose, hemicellulose and lignin content) can predict the heating value of the biomass fuel (Mitchell et al., 2003). Collectively, these studies point out the great variability in physical and/or chemical characteristics observed between available biomass resources, and the need to utilize sources with defined characteristics that are most appropriate to the desired application. The identification of specific measurable properties that correlate significantly with increased application

efficiency will assist in matching high volume agricultural by-products, such as almond shells, with their most appropriate industrial uses.

We have evaluated eight almond accessions in this study and found significant variability in shell cracking strength, even among almond accessions with non-significant differences in kernel percentage. The observed variability of this physical character, even on a limited number of accessions, demonstrates that this large volume by-product of the almond industry is not homogeneous. To date, studies involving potential value-added uses of almond shells have been non-specific with regard to the varietal designation of the shells being used. Recent investigations have determined that the suitability of various agricultural wastes for activated carbon production is not material specific, but is determined by the particular waste's specific physical and chemical properties, as well as the conditions under which the carbons are produced (Aygün et al., 2003). Certain almond cultivars may therefore have ideal properties that make them particularly suitable for specific value-added processes. Hence, future research should sample these and other major tonnage almond varieties (Butte, Carmel, Nonpareil, etc.) for other important physical and chemical characters, as well as produce batch-sized volumes of carbons from the shells of specific cultivars for product comparisons. Systematic evaluation of almond varieties and characterization of their shell materials will be an important and necessary step in determining the best value-added uses are for this abundant biomass.

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